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Intercontinental transport of anthropogenic sulfur dioxide and other pollutants: An infrared remote sensing case study

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[1] Using 3 years worth of IASI (the Infrared Atmospheric Sounder Interferometer aboard METOP-A) measurements, we have identified 24 major events of uplift and transport of anthropogenic sulfur dioxide. These were all first observed over East Asia, and could be traced for over 60 hours. On 7 November 2010 a sulfur dioxide plume was observed over Northeast China and tracked for five days to North America. We discuss this event in detail with respect to build up; uplift and in-plume chemistry. We found a host of trace gas enhancements in the plume (SO_2 , CO, PAN, CH_3OH , HCOOH and C_2H_2). A reasonable to very good agreement was found with MOZART-4 modeled ambient columns for all species except methanol, which was underestimated by the model by an order of magnitude. We calculate correlations of the different species and give observational evidence of secondary in-plume formation of methanol and PAN.
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1. Introduction

[2] Transport of pollution has an impact on atmospheric composition, chemistry and air quality across continents from the boundary layer up to stratosphere. Its importance has been established through a large number of model studies and in situ measurements [see, e.g., Liang *et al.*, 2004; Heald *et al.*, 2003; Talbot *et al.*, 2003; Kritz *et al.*, 1990; Chin *et al.*, 2007; Stohl, 2001; Fiedler *et al.*, 2009; Barrie, 1986; Law and Stohl, 2007; Jaffe *et al.*, 1999]. Outflow from East Asia is largely controlled by monsoons and throughout the year reaches North America either via the North Pacific or via the Arctic (mostly in the winter) [Stohl *et al.*, 2002]. Uplift and export mechanisms have a strong seasonal variability with spring being the most favorable period for transpacific transport due to active weather associated with frequent ascending air parcels and

fast moving westerly winds [Liu *et al.*, 2003; Russo *et al.*, 2003].

[3] The IASI infrared sounder [Clerbaux *et al.*, 2009] aboard MetOp-A has twice a day global coverage, which allows detection and analysis of the evolution of atmospheric plumes such as those arising from a volcanic eruption [Clarisse *et al.*, 2008] or biomass burning [Coheur *et al.*, 2009]. Here for the first time we use IASI to study transport of anthropogenic pollution plumes and analyze a particular plume first spotted above East Asia on 7 November 2010. IASI has a good spectral resolution, which permits measurement of a variety of trace gases [Clarisse *et al.*, 2011]. For this event we were able to measure plume enhancements of carbon monoxide (CO), sulfur dioxide (SO_2), peroxyacetyl nitrate (PAN), formic acid (HCOOH), acetylene (C_2H_2), methanol (CH_3OH) and their evolution in time as the plume diluted and underwent chemistry and washout. It is the first study of this type as satellite studies of Asian anthropogenic outflow have traditionally been focused on CO [Turquety *et al.*, 2008; Zhang *et al.*, 2006] and aerosols [Yu *et al.*, 2008; Di Pierro *et al.*, 2011]. Infrared sounders are generally not sensitive to anthropogenic SO_2 ; and we therefore start section 2 by giving an overview of the frequency and seasonality of such anthropogenic SO_2 observations, before describing the event observed in November 2010. In section 3 we give a quantitative analysis of the event for the reported trace species and briefly compare with outputs from the MOZART model.

2. Anthropogenic SO_2 and the November Event

[4] While UV-vis instruments routinely measure anthropogenic SO_2 [Lee *et al.*, 2011], this is not the case for infrared instruments which have limited sensitivity to boundary layer SO_2 . For the ν_3 absorption band of SO_2 (which is the strongest absorption band in the infrared) this is due to competing, and dominating water vapor absorption in the same spectral range. When SO_2 is observed in this band it must be present in a significant amount at altitudes above the lower troposphere (5–7 km depending on the water vapor profile and SO_2 concentrations [Clarisse *et al.*, 2008]).

[5] We have analyzed SO_2 retrievals for the complete period October 2007–December 2010 in search of anthropogenic emissions of SO_2 . Because of frequent volcanic activity, especially in the Northern Hemisphere, the produced imagery was analyzed manually and cross checked with volcanic activity reports from the USGS (<http://www.>

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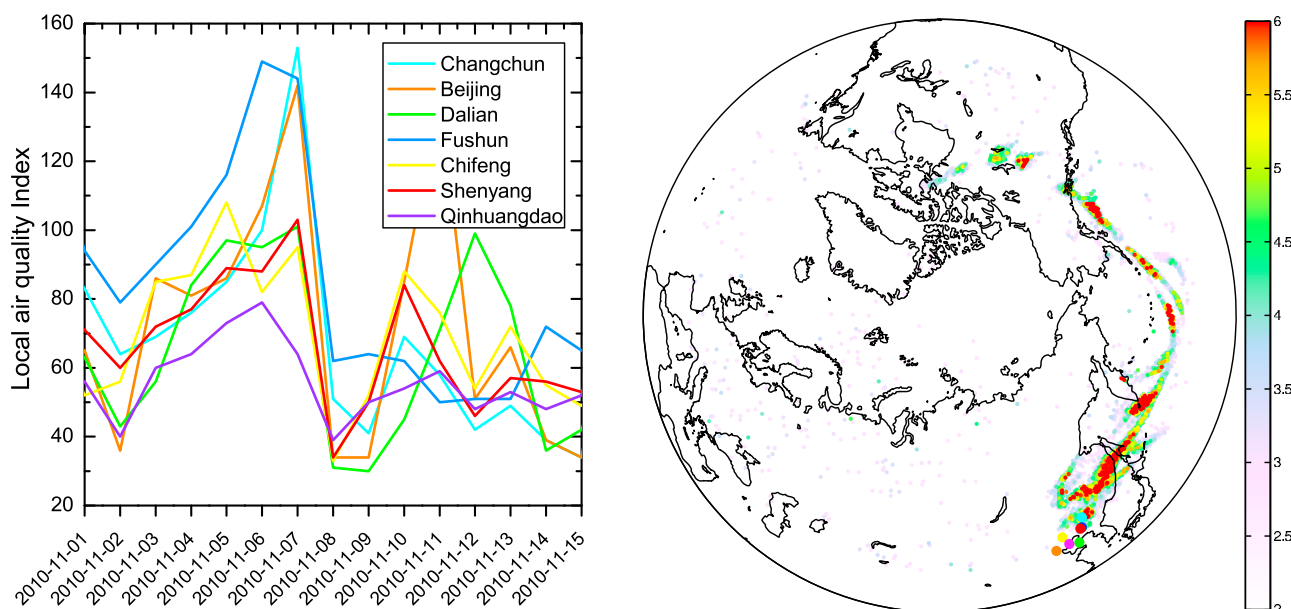


Figure 1. (left) Daily Air Pollution Index from selected cities in China. The air quality decreases from 1 to 7 November and is attributed to build-up of pollution. On the 8th a drastic improvement is apparent, coincident with the IASI observation of the SO_2 plume shown on the right. (right) Location of selected cities (colored dots) and maximum observed SO_2 columns (in units 10^{16} molecules/ cm^2) for the period 7–11 November 2010.

volcano.si.edu/reports/usgs/). In total we identified 24 major events where an apparent non-volcanic SO_2 plume was observed with loadings above 5 km exceeding $\sim 5 \cdot 10^{16}$ molecules/ cm^2 and which was visible for at least 5 consecutive IASI overpasses (2½ days). All these were first observed over East Asia and Siberia; while transport was mainly observed over the Russian Arctic, Japan, Sakhalin and Kamchatka. There is a marked seasonality in the events with 12 events observed in the fall, 9 in the winter and 3 in the spring.

[6] A first prerequisite for such large events to happen and for infrared sounders to see them, is the availability of large concentrations of SO_2 . Note first that emissions of SO_2 only show a weak seasonal pattern [Streets *et al.*, 2003]. In colder periods, the lifetime of SO_2 is however longer (a factor 2–3 in winter [Chin and Jacob, 1996; Lee *et al.*, 2011]) because of slower dry deposition and less availability of oxidants. Furthermore, SO_2 is half-soluble and subject to wet scavenging. Over East Asia, precipitation has a marked seasonality, with fall and winter being very dry and spring and especially summer very wet. Enhanced with trapping of boundary layer air due to inversion, fall and winter are therefore favorable periods for the build up of large concentrations of SO_2 . The second prerequisite is an efficient uplift mechanism to altitudes above 5 km. The occurrence of midlatitude cyclones and the associated warm conveyor belts are the main source of uplift to the mid-upper troposphere and occur frequently in fall and winter (note that these occur less frequent in the summer) [Stohl, 2001; Liang *et al.*, 2004; Yienger *et al.*, 2000].

[7] This seasonality is very different as compared to CO, which is known to exhibit most transpacific transport in the spring (and to lesser extent in the summer). This can be seen e.g. from looking at monthly averages of CO [Clerbaux *et al.*, 2009, Figure 5]. To understand this apparent contradiction, it is important to take into account the seasonality in

the number of uplift events, the availability of source material and the altitude of transport. In the work of Liang *et al.* [2004] it was shown that the number of uplift events does not exhibit a strong seasonality (although the uplift mechanism can be different, e.g. in summer convective lifting is more important). However, in spring and summer, the amount of CO that is lifted and transported is larger due to larger source availability. Secondly, here we have only taken into account SO_2 transport events above 5 km, while the well known seasonality of CO transport includes transport at all altitude levels. Finally, the number of large uplift events might not be a good indication of the total amount of transported material, for which also a large number of smaller events can contribute.

[8] On 7 November 2010, IASI observed a SO_2 plume covering almost the whole of Northeast China and subsequent overpasses show the plume moving over the Sea of Japan, Northern Pacific, Gulf of Alaska and Canada. The last observations was made 5 days later near Hudson bay (see Figure S1 and Animation S1 in the auxiliary material).¹ Fortuitous overpasses of the space-borne lidar Calipso [Winker *et al.*, 2009] revealed coincident aerosol features between 6 and 9 km which were identified as smoke and in the later stages as sulfate. For all but the first overpass there was part of the aerosol curtain which contained no detectable clouds, and these were selected for Figure S1 in the auxiliary material.

[9] In terms of meteorology, the week prior the event was characterized by low surface winds and a total absence of rain. These stagnant conditions led to build up of pollutants as reflected in the steady increase of the Daily Air Pollution Index [Gao *et al.*, 2011] of several megacities in Northeast China during that week (see Figure 1, left). Weather maps of

¹Auxiliary materials are available in the HTML. doi:10.1029/2011GL048976.

Table 1. Overview of the event^a

Day	Clouds (%)	Size (km ²)	CH ₃ OH	HCOOH	CO	SO ₂	PAN	C ₂ H ₂
07AM	-	-	0.30e16	0.55e16	2.00e18	5.00e16 (25)	0.40e16 (1.5)	0.75e16 (3.0)
07PM	73	0.6e6	3.17e16	1.00e16	1.65e18	5.93e16 (25)	0.75e16 (3.8)	1.00e16 (4.7)
08AM	61	1.0e6	3.91e16	1.25e16	1.80e18	6.27e16 (21)	1.04e16 (3.6)	1.05e16 (4.3)
08PM	47	1.4e6	4.04e16	1.55e16	2.01e18	6.46e16 (26)	1.69e16 (4.0)	1.12e16 (4.1)
09AM	34	1.6e6	3.42e16	1.26e16	1.58e18	5.42e16 (25)	1.13e16 (4.7)	0.82e16 (3.1)
09PM	36	1.1e6	3.16e16	1.20e16	1.58e18	5.17e16 (24)	1.05e16 (3.6)	0.80e16 (2.6)
10AM	36	1.1e6	3.20e16	1.03e16	1.21e18	4.87e16 (26)	0.81e16 (3.7)	0.64e16 (3.1)
10PM	17	0.7e6	2.84e16	0.99e16	1.23e18	4.55e16 (28)	0.63e16 (3.1)	0.72e16 (2.4)
11AM	7	0.4e6	2.74e16	0.94e16	0.70e18	4.00e16 (22)	0.48e16 (2.8)	0.57e16 (2.7)
11PM	8	0.3e6	2.69e16	0.76e16	0.92e18	3.59e16 (29)	0.38e16 (0.8)	0.49e16 (2.3)

^aFirst column is the date (in November 2010); and the overpass (morning or evening). The second column is the average cloud coverage from the IASI level 2 data. The third column is the size of the plume as estimated from the SO₂ retrievals. The remaining columns are maximum observed column enhancements (averaging the 10 highest observations) in molecules/cm². For the species which correlate well with CO the enhancement ratio or slope $\Delta X/\Delta CO$ is shown in brackets in units of 10⁻³. The first line (07AM) represent maximum MOZART4 modeled columns below 800 hPa over the likely source area prior to the event.

the 6th and the 7th November show an intensifying baroclinic cyclone over Northeast China and Korea forming thick clouds and accompanying rain further northeast of this area. The associated warm conveyor belt is no doubt responsible for the lofting and entraining of the local pollutants picked up by IASI. Note that associated cloud formation is important because of potential washout of pollutants and the remote sensing challenge of dealing with optical thick clouds. These can hamper the retrieval and, as we will see further, bias it low.

3. Composition, Columns and Comparisons

[10] Comparison of SO₂ data with near real time CO retrievals [George *et al.*, 2009] reveal coincident enhancements. To assess enhancements of other species in the plume, differences of averaged spectra inside the plume (using spectra with SO₂ columns above $\sim 5 \cdot 10^{16}$ molecules/cm²) and outside the plume (spectra with no detectable SO₂) were made. This technique allows detection of weak absorbers, as it reduces noise and removes a large part of the spectral signatures due to the strong absorbers (CO₂, O₃, H₂O [Karagulian *et al.*, 2010]). Unambiguous identification of CO (3 K), PAN (0.4 K), HCOOH (0.5 K), C₂H₂ (1 K), CH₃OH (0.4 K) was possible for each overpass. Here the numbers in brackets are averaged observed signal strengths and these emerge well above the IASI noise (~ 0.2 K on an individual spectrum).

[11] For all detected species, we have performed optimal estimation retrievals using Atmosphit [Coheur *et al.*, 2005, 2009], limiting the retrieval of the target species between 6 and 9 km (except for CO which was fitted from 0 to 15 km). Cloudy scenes are problematic since clouds are not directly taken into account by Atmosphit. Thin clouds or small cloud fractions can be compensated by a correction in surface temperature, but thick clouds distort the baseline shape and lead to diverging fits. For this reason we have performed optimal estimation retrievals only for spectra with a cloud coverage below 10%. Unfortunately, especially for the first overpasses, the cloud fraction was very high (73% clouds on average for the first overpass, see Table 1), resulting in only few retrievals, mostly on the plume edge (and thus missing the highest loadings). Recently, Walker *et al.* [2011] introduced a method of detecting weakly absorbing species yielding an apparent column amount. For fixed atmospheric conditions and far away from saturation, the apparent col-

umn is linearly related with the true column amount. We have implemented the detection using the so-called ensemble method from Walker *et al.* [2011] and confirmed a strong linear correlation (between 0.75 and 0.95) between apparent and observed columns. This was used to obtain true column amounts for all pixels in and around the observed SO₂ plume. This method does not work for CO, which is a strong absorber with a saturating spectral signature and omnipresent in the atmosphere.

[12] The retrievals obtained in this way are illustrated for SO₂, HCOOH, C₂H₂ and PAN in Figure 2 and summarized in Table 1, which contains maximum measured columns (mean of the largest ten observations). For CO, mean out-of-plume columns were subtracted from the total columns and as the other species were not detected outside the plume we can interpret all columns as in-plume enhancements. For all species maximum loadings were detected on the third overpass (evening overpass of the 8th), indicating underestimated loadings for the first two overpasses due to cloud coverage.

[13] By inter-comparing the different species, more can be said on the in-plume chemistry and lifetime. CO and C₂H₂ abundances are usually correlated in the atmosphere [Xiao *et al.*, 2007]. Because of difference in lifetime, the ratio C₂H₂/CO or enhancement ratio $\Delta C_2H_2/\Delta CO$ can be used as a tracer of air mass age. We found an average correlation coefficient of 0.59. The initial value for the enhancement ratio was found to be $4.7 \cdot 10^{-3}$, with the slope declining to $2.3 \cdot 10^{-3}$ for the aged plume. These values were obtained from linear regression of all measurements in the plume. The initial value matches closely the reported emission average of $4.8 \cdot 10^{-3}$ [Xiao *et al.*, 2007] for East Asia.

[14] The best correlation with CO was found with PAN with an average correlation coefficient of 0.68. PAN production peaks in the summer, however photochemical production of PAN can also be elevated outside the summer and thermal stability at lower temperatures can account for increased concentrations [Brice *et al.*, 1988; Tsalkani *et al.*, 1987]. Its enhancement ratio with CO was initially $\sim 3.7 \cdot 10^{-3}$, increased to $\sim 4.7 \cdot 10^{-3}$ and then dropped the last day to $\sim 0.8 \cdot 10^{-3}$. On the third overpass remarkably high columns were measured (more than a factor two of what was observed during the first overpass). This increase is much larger than any of the other species, and therefore unlikely related to the cloud retrieval problem. It is indicative of secondary formation within the plume.

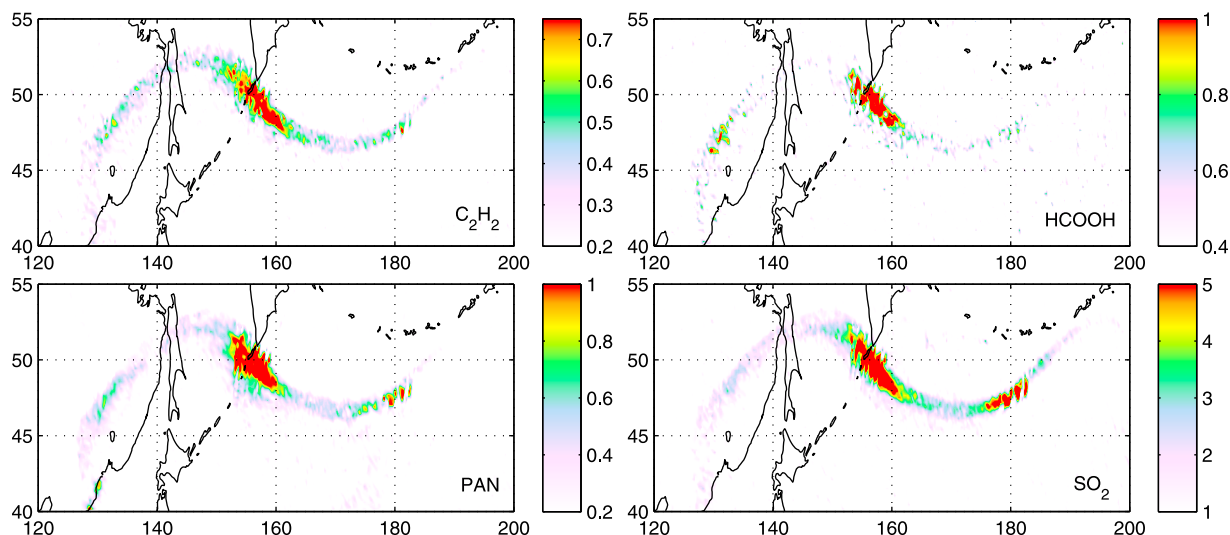


Figure 2. Example retrievals on 8 November 2011, PM of C_2H_2 , SO_2 , PAN and HCOOH in units 10^{16} molecules/ cm^2 . While the abundances of these species correlate well (see also Figure S2 in the auxiliary material), the shape and location of the plume match almost perfectly. It can also be noted that HCOOH is only barely detectable in parts of the plume.

[15] All four species: PAN, C_2H_2 , CO and SO_2 correlated well with each other throughout the event, with correlation coefficients above 0.5. Correlations with HCOOH and CH_3OH were lower (in the range 0.2–0.5). This is mostly due to the larger noise of their retrievals (see Figure 2). Formic acid and methanol are emitted biogenically, with production peaking in the growing season, but also by fires, combustion and other anthropogenic activities [Stavrakou *et al.*, 2011]. The main sink of formic acid is wet deposition, and concentrations are hence strongly linked with rainfall [see, e.g., Zhang *et al.*, 2011]. As there was no precipitation in the week building up to the event, it is not surprising that this species was observed here. Of all the observed species, methanol abundances have the slowest decrease rate from the third to the last overpass, see Table 1 (slower than SO_2 or the long-lived CO). Since dispersion of the plume is the same for all observed species, and since dispersion is the main reason for the observed decrease in CO abundances, the slower decrease of CH_3OH is strongly suggestive for in-plume formation, probably related to the oxidation of volatile organic compounds.

[16] We have also carried out a comparison of our measured columns with MOZART-4, driven by NCEP/GFS meteorology (see <http://www.acd.ucar.edu/acresp/forecast/> for the figures), in order to see how a state of the art global chemistry transport model deals with such an event of strong uplift and long range transport. From the CO maps, outflow to the Pacific is obvious on the 8th of November. Transport is much slower though, as by the 11th the plume is still modeled over the Western Pacific, because of an underestimation of uplift and altitude. This is not surprising as modeling uplift and subsequent circulation through moist adiabatic storms is highly non-trivial. To assess the chemistry modeling of the build up, our measured columns have been compared with maximum measured columns below 800 hPa in the morning of the 7th over the likely source area. Modeled columns agree well for CO, C_2H_2 and SO_2 but are underestimated for formic acid and PAN (almost a factor two) and especially methanol (a factor ten). These species are not surprisingly those which are least monitored

(e.g. to our knowledge there has not been a single recent in-situ campaign aimed at measuring wintertime PAN in North East Asia).

[17] This study provides a detailed analysis of one of the 24 identified events of major uplift and transport of anthropogenic sulfur dioxide. It is a first demonstration of how IASI is able to monitor outflow and chemistry of a range of primary and secondary pollutants. As retrieval techniques and instrumental characteristics of infrared sounders continue to improve, we expect it to be possible to monitor outflow of Asian pollution of a series of trace gases on a more regular basis, offering the modeling community unprecedented validation opportunities.

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